An Investigation of the Impact of Canadian wildfires on US Air Quality using ~~Satellite,~~ Model, Satellite and Ground Measurements

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Abstract.

Canadian wildfires transport large concentrations of particulate matter into the United States, leading to ~~changes in~~ various impacts on surface temperature, radiation balance, visibility, and exacerbating pollution-related respiratory conditions. Using a combination of surface, satellite and numerical models, this study quantifies the increase in surface fine particulate matter (PM2.5) in the Continental United States due to long-range transported smoke from Canadian wildfires during a wildfire episode from August 9th to 25th, 2018. As a widely used indicator of surface pollution levels, satellite-retrieved AOD can provide ~~vital~~ crucial information on columnar pollution mass. However, the daily spatial coverage of satellite AOD is restricted due to cloud cover. In order to quantify the daily changes of surface pollution, we fill in the AOD gaps by utilizing simulated ~~10-m~~ 10-km spatial resolution AOD from a chemistry transport model (CTM). ~~In addition, different processes affecting smoke vertical and horizontal transport are examined using two CTM simulations. Meteorological variables associated with these processes are then selected along with~~ Meteorological variables influencing smoke transport were also integrated alongside the gap-filled AOD product to ~~assess the surface pollution~~ estimate surface PM2.5 using geographically weighted regression (GWR) and random forest (RF) models. ~~It is found that synoptic pressure systems dominate the horizontal transport of Canadian smoke, and the lifting of aerosols from a low-pressure system contributes to the long distances of the transport path. Other processes affecting the vertical distribution of pollutants, including boundary layer entrainment, precipitation and terrain-induced vertical mixing are also analyzed in this paper~~The model with better performance was subsequently applied to quantify PM2.5 changes due to Canadian wildfires. To isolate the impact of Canadian wildfires, we calculate the surface PM2.5 ratio with and without Canadian fire sources by conducting two CTM simulations: one with Canadian wildfire emissions enabled and another with these emissions turned off. Our results show that Canadian ~~fires increase total~~ wildfires caused a significant increase in surface PM2.5~~up to 13% (62~~, contributing up to 28 μgm^{-3} ~~) over~~ (a 69% increase) across different US EPA regions during ~~2018 August~~ the August 2018 wildfire event.

1 Introduction

Airborne fine ~~particle mass concentrations~~ particulate matter (PM2.5), with aerodynamic diameters less than 2.5μm~~(~~, is a well-documented contributor to increased mortality from diseases such as ischemic heart disease, chronic obstructive pulmonary

disease, cardiovascular disease, respiratory disease, lung cancer, chronic kidney disease, hypertension, and dementia (Chen and Hoek, 2020 In 2017, PM2.5 ~~) exposure contributed~~ exposure was linked to 4.58 million deaths globally~~in 2017, and~~ , with ambient PM2.5 ~~were responsible~~ accounting for 64.2% of these deaths (Bu et al., 2021). PM2.5 ~~typically originates from combustion sources,~~ originates from diverse sources, including combustion processes, power plants, dust, sea salt, and secondary chemical reactions~~, and wild-land fires are primary sources~~ . In the United States, wildfires are a significant and growing source of PM2.5 ~~in the US~~ pollution (O'Dell et al., 2019). For regions affected the most by wildfires, like ~~Washington state~~ the state of Washington (WA), an increase in daily PM2.5 of 97.1 μgm^{-3} during the summer of 2020 was found, which related to 92 more mortality cases (Liu et al., 2021). Moreover, the toxicity of PM originating from wildfire smoke ~~is~~ has been found to be 3-4 times greater than equivalent doses of ambient PM (Wegesser et al., 2009). Health impacts vary ~~depending upon~~ with chemical composition, which ~~in turn varies~~ depends upon the stages of biomass combustion and combustion depends on biomass combustion stages and temperature (Kim et al., 2018; Aguilera et al., 2021). ~~Previous studies show clear linkages between PM2.5 and increased mortality from diseases including Ischemic heart disease, Chronic obstructive pulmonary disease, cardiovascular disease, respiratory disease, lung cancer, chronic kidney disease, hypertension, and dementia (Chen and Hoek, 2020; Bu et al., 2021; Bowe et al., 2019). The association exists not only for high levels of PM2.5 concentrations but also for the WHO guideline exposure level of 10 μgm^{-3} (Chen and Hoek, 2020). Besides the~~ Beyond its health impacts, ~~wildfires' increasing economic impacts have become another burden on the community~~wildfire-related PM2.5 imposes considerable economic burdens. From simulation results, wildfire-related economic costs ~~increased~~ have been projected to increase from $ 7 billion per year to $ 43 billion per year in 2090 (Neumann et al., 2021).

Given the growing role of wildfires as a major PM2.5 source, accurately assessing pollution levels requires understanding the transport dynamics and chemical transformations of wildfire smoke, which are influenced by factors such as fire intensity, injection height, atmospheric dynamics, and terrain interactions. Higher fire radiative power (FRP) results in longer distances of the smoke transport due to higher ~~injection height of the plumes (Solomos et al., 2015), and the processes that bring these injected aerosols to the surface can be complex. Injection heights vary for different fires, and it can reach 15km for certain tropical fires (Paugam et al., 2016). For a Canadian wildfire in 2002, the injection height was estimated to be 2-6 km to match the lidar observed vertical profile from the Micro-Pulse Lidar NETwork (MPLNET) (Colarco et al., 2004). The long-range transported elevated layers of smoke from Canadian forest fires gradually subsided~~ plume injection heights (Solomos et al., 2015). A global analysis of over 23,000 wildfires found that significant injection heights into the free troposphere are primarily observed in the boreal forests of North America and Siberia during the northern summer (Val Martin et al., 2018). Smoke remnants from Canadian stand-replacing forest fires have been observed at altitudes exceeding 13 km (Damoah et al., 2006). Once injected, smoke plumes can descend to the surface ~~in the Eastern US through combinations of subsidenceprocess~~through a combination of subsidence, interception, and ~~subsequent entrainment process by the diurnal variations of Planetary boundary layer~~ diurnal entrainment within the Planetary Boundary Layer (PBL)~~(Colarco et al., 2004)~~, as observed in the Eastern US for 2002 July Canadian forest fire event (Colarco et al., 2004). Atmospheric circulation plays a key role in shaping smoke transport. ~~Synoptic scale pressure system controls the transport of smoke from its sources.~~ Upper-level winds ~~transport the smoke horizontally, and surface high pressure is found to be related to surface pollution increase~~ facilitate long-range horizontal

transport, while surface high-pressure systems enhance ground-level pollution through subsidence inversions (Miller et al., 2011). Cyclonic circulation can form a multilayer PBL~~which is usually associated with temperature inversion~~ , characterized by temperature inversions and stable stratification, ~~causing accumulation of pollution in the convergent zone~~ which trap pollutants in convergent zones (Jiang et al., 2021b). Interactions ~~between mountain terrain with synoptic high-pressure systems enhance the stability in valleys (Beaver et al., 2010), while the same terrain can also enhance vertical mixing under unstable synoptic conditions (Lang et al., 2015). During the transportprocess, various processes may also change aerosol properties. For example, thick black carbon (BC)-free coatings with BC cores are found for long-range transport smoke particles~~ with mountain terrain further modulate smoke dispersion: under stable synoptic conditions, valleys become more stagnant, while unstable conditions promote vertical mixing (Beaver et al., 2010; Lang et al., 2015). During transport, ~~which increase the light absorptivity of the aged smoke (Dahlkötter et al., 2014).~~

~~Aged smoke particles can grow larger via various mechanisms, including hygroscopic growth~~ smoke particles undergo physical and chemical transformations that influence their sizes, such as hygroscopic growth (Carrico et al., 2005; Gomez et al., 2018), SOA formation (Ahern et al., 2019), condensation of semi-volatile species (Reid et al., 2005; Zhou et al., 2017; Akagi et al., 2012), ~~condensation of volatile organic species ,~~ and coagulation process ~~(Colarco et al., 2004; Müller et al., 2007), and the larger sizes may enhance inflammogenic and cytotoxic activities of the particle (Jalava et al., 2006). Although significant increases in PM2.5 were found for long-range transport wildfire smoke, the toxicity of organic compounds may be reduced during the transport process (Jalava et al., 2006). Some studies found long-range transport PM positively associated with cardiovascular mortality over thousands of kilometers from the sources (Kollanus et al., 2016; Magzamen et al., 2021). In contrast, some found no relation between short-term elevation in PM2.5 and daily mortality (Zu et al., 2016).~~ (Aloyan et al., 1997; Sun et al., 2019).

~~In order to assess the surface pollution levels, different approaches have been used for estimating~~ Estimating surface PM2.5 concentrations ~~: spatial interpolation (inverse distance method~~presents significant challenges due to spatial and temporal variability and limited ground station coverage. Traditional approaches, such as spatial interpolation (e.g., inverse distance weighting, ordinary kriging~~method) , linear regression (Hoff and Christopher, 2009),)~~ rely primarily on ground-based PM2.5 measurements, while linear regression methods combine satellite-derived AOD with surface PM2.5 measurements (Hoff and Christopher, 2
More advanced methods, like multi-linear regression~~(Gupta and Christopher, 2009b), geographically~~ , typically incorporate surface PM2.5 data, satellite AOD, and meteorological datasets from models (Gupta and Christopher, 2009b). Geographically weighted regression (GWR) ~~(Xue et al., 2021; Ma et al., 2014; Bai et al., 2016; Song et al., 2014), linear~~ and machine learning methods use similar data sources as multi-linear regression but offer better spatial resolution and adaptability to local variations (Xue et al., 2021; Ma et al., 2014; Bai et al., 2016; Song et al., 2014; Hu et al., 2017; Gupta and Christopher, 2009a; Zamani Joharestani e
Linear mixed-effect ~~model (Ma et al., 2016; Lee et al., 2011), chemistry transport model (CTM) methods (Geng et al., 2015; Xue et al., 201 machine learning models (Hu et al., 2017; Gupta and Christopher, 2009a; Zamani Joharestani et al., 2019). Traditional approaches are outdated as newer techniques combine different mechanisms and add various variables with~~ models add temporal variability by including both fixed and random effects (Ma et al., 2016; Lee et al., 2011). While chemistry transport models (CTMs) leverage detailed atmospheric chemistry and physics simulations using emissions inventories, meteorological fields, and chemical

species distributions (Geng et al., 2015; Xue et al., 2019). Traditional methods like spatial interpolation and linear regression struggle to integrate multiple mechanisms and spatial-temporal ~~information to improve their prediction accuracy Zhang et al. (2018)~~variable a limitation newer techniques address (Zhang et al., 2018). Due to the growth of computing power, machine learning (or artificial intelligence) has become a major focus for estimating the spatial-temporal dynamic distribution of surface PM2.5 concentrations ~~Zhang et al. (2018); Sayeed et al. (2022)~~(Zhang et al., 2018; Sayeed et al., 2022).

~~Satellite retrieved AOD is an essential indicator of surface pollution level (Wang and Christopher, 2003; Geng et al., 2015). These data are beneficial because more than a 20-year record of satellite AOD is available~~ Satellite-retrieved AOD serves as a valuable indicator of columnar pollution, providing critical information for estimating surface PM2.5 concentrations, though it is not a direct measurement of surface pollution (Wang and Christopher, 2003; Geng et al., 2015). Its key advantages include over two decades of data availability from polar-orbiting sensors and the extensive spatial coverage of satellite observations, making it a powerful tool for long-term air quality analysis. However, cloud cover ~~obstruct AOD retrieval and thus affect~~ frequently obstructs AOD retrievals, limiting their utility for daily surface PM2.5 ~~estimations on a daily basis (Goldberg et al., 2019). Therefore, gap-filled AOD values can provide more useful information on surface pollution variations and distribution because most techniques that use satellite AOD to estimate surface PM2.5 require reliable data at each pixel. Fusion of AOD retrievals from different satellite sensors largely improves the AOD coverages Ma et al. (2014). Another way is to use multiple imputation methods with inputs including~~ estimation (Goldberg et al., 2019). To overcome this limitation, gap-filling methods are used to generate spatially and temporally continuous AOD datasets. These methods include fusing retrievals from multiple satellite sensors (Ma et al., 2014), applying multiple imputation techniques using auxiliary data such as cloud fraction, elevation, ~~humidity, temperature, and spatiotemporal trends to impute the missing AOD values (Xiao et al., 2017). Based on the linear relationships of AOD and~~ and humidity (Xiao et al., 2017), and using statistical approaches like Kriging to interpolate AOD based on seasonal and regional AOD:PM2.5 ~~, AOD values estimated using seasonal, regional-specific coefficients derived from surface PM2.5 measurements can be then interpolated (Kriging) to obtain a more extensive coverage AOD product Lv et al. (2017). Comparing various~~ ratios derived from ground-based measurements (Lv et al., 2017). Additionally, incorporating chemistry transport model (CTM) simulations into the gap-filling ~~methods, the inclusion of CTM simulations essentially improves the accuracy of gap-filling results Xiao et al. (2021)~~process has been shown to significantly improve the accuracy and reliability of imputed AOD values (Xiao et al., 2021). The AOD-PM2.5 relationship is influenced by aerosol composition, vertical distribution, and meteorological factors. Planetary Boundary Layer Height (BLH) serves as a key indicator by representing the mixing height of aerosols and their influence on surface pollution levels (Gupta and Christopher, 2009b). Surface pressure also plays a role, with high-pressure systems linked to surface pollution increases due to subsidence inversions (Miller et al., 2011). Additionally, Relative Humidity (RH) affects this relationship, as AOD is sensitive to particle size and hygroscopic growth (Li et al., 2014), while PM2.5 reflects dry mass. Incorporating these meteorological parameters helps models better capture the dynamic processes shaping the AOD-PM2.5 relationship.

This study is designed to assess the pollution change due to fine particulate matter in the United States transported by smoke from Canadian wildfires and to also examine the physical processes during transport that affects surface pollution along the path. First, by turning on and off the fire emissions in the CTM in Canada, we conduct two WRF-Chem simulations to in-

vestigate the processes that influence the transport of remote smoke aerosols in the atmosphere. According to the analysis of different processes, we then selected variables associated with smoke''s vertical distribution, which determined the relationships between AOD and surface pollution concentrations. Finally, by filling in the satellite AOD gaps due to cloud covers using CTM simulations, we estimate the surface pollution increase due to the remote fires using the filled AOD along with surface PM2.5 measurements and other meteorological variables.

2 Data and Study Area

2.1 Study area

We estimated daily mean surface PM2.5 at 0.1-degree spatial resolution over Continental US (CONUS) from August 9th to 25th, 2018. The study area ~~is focused~~ (inner domain) focuses on the US (25-50° N, 64-125° W)~~. At the same time, we also include the Canada region~~ , while the outer domain of the same WRF simulation extends to Canada (25-67° N, 70-140° W) ~~when performing the WRF simulation~~ to account for ~~the fire emissions in Canada to investigate the influence of pollution within the US caused by~~ Canadian fire emissions and their contributions to US pollution from remote fire sources. Therefore, we chose August 2018 as our study period to analyze the impact of wildfires on surface air pollution in the US based on the total fire radiative power ~~calculations~~ calculation based on our previous work (Xue et al., 2021).

2.2 Ground-level PM2.5 observations

We obtained the daily surface PM2.5 concentration product that uses ~~federal reference methods (FRM, with codes~~ Federal Reference Methods and Federal Equivalent Methods (FRMs and FEMs, with code 88101) from US Environmental Protection Agency (EPA) for CONUS within the study period (https://www.epa.gov/outdoor-air-quality-data/). The measured frequency of each site is different (with a measurement interval of every 1, 3 or 6 days). A total of 950 EPA sites are available with approximately 71.1% sampling for daily data within the study period. Note that we discarded all PM2.5 values lower than the established detection limit of 2 $\mu g m^{-3}$ (EPA, 2018).

2.3 Satellite data

AOD values retrieved from satellite observations which is a columnar value of aerosol extinction, are correlated with surface pollution under certain conditions (Wang and Christopher, 2003; Hoff and Christopher, 2009). The relation between AOD and surface PM2.5 can be expressed as the following equation (Koelemeijer et al., 2006):

$$AOD = PM_{2.5} H f(RH) \frac{3 < Q_{ext,dry} >}{4 \rho r_{eff}} \quad (1)$$

From the above equation, several factors that bridge AOD and PM2.5, including: aerosol layer height H, particle effective radius r_{eff}, aerosol mass density ρ, extinction efficiency under dry conditions $Q_{ext,dry}$, and the ratio of ambient and dry

extinction coefficients f(RF). Thus satellite AOD retrievals are often used as a vital indicator for estimating surface pollution (Hu et al., 2014; Xie et al., 2015).We use the 550nm AOD from the Multi-Angle Implementation of Atmospheric Correction (MAIAC MCD19A2 collection 6 product) with 1-km spatial resolution (https://lpdaac.usgs.gov/products/mcd19a2v006/). The product retrieves AOD from Terra and Aqua MODIS (Moderate Resolution Imaging Spectroradiometer), and we take the mean value of all available retrievals. Considering that thick smoke is likely misclassified as clouds, we accept all AOD with or without adjacent clouds (Xue et al., 2021; Goldberg et al., 2019). Through ~~validating~~ validation with AERONET AOD, the accuracy of the MAIAC AOD product is approximately 66% within the expected error (± 0.05 ± 0.1 AOD) (Lyapustin et al., 2018). The 1-km resolution AOD is gridded to 10 km by averaging all valid AOD values in 0.1-degree boxes. It is worth noting that MAIAC AOD is originally retrieved at 470nm and then ~~computed~~ computes the 550nm AOD using spectral properties (Lyapustin et al., 2018; Liu et al., 2019). It has also been reported that the uncertainties and biases increase with increasing AOD (Martins et al., 2017; Qin et al., 2021).

2.4 Meteorological data

The AOD-PM2.5 relationship depends on various factors, including meteorological parameters (Xue et al., 2021). Boundary layer height (BLH), 2-meter temperature (T2M), 10-meter wind speed (U10M), surface relative humidity (RH), and surface pressure (SP) were obtained from the European Centre of Medium-Range Weather Forecasts (ECMWF) Re-analysis (https://www.ecmwf.int/en/forecasts). To match the AOD data with an average value of two different times (10:30 am and 1:30 pm), we downloaded all meteorological variables at 12 pm local time. The spatial resolution of all meteorological data is 0.25 degrees, and we use the inverse distance method to interpolate all the variables to 0.1-degree spatial resolution.

3 WRF-Chem model and experimental design

The Weather Research and Forecasting model coupled with Chemistry (WRF-Chem V4.2.2) is applied in this study to examine the various processes that affect the transport of smoke aerosol and estimate the pollution change in the US due to Canadian fires. This section briefly describes the WRF-Chem model, the model configuration, and model physics and then introduces the design of the numerical experiments.

3.1 WRF-Chem model

WRF is a state-of-art mesoscale numerical weather prediction system that offers operational forecasting a flexible and computationally-efficient platform (Skamarock et al., 2019). WRF-Chem is an ~~in-line~~ atmospheric chemistry model ~~(Powers et al., 2017)~~ that fully integrated with the meteorological framework of WRF (Powers et al., 2017), enabling the simulation of various chemical and physical processes related to aerosol transport, including dispersion, aerosol-cloud interactions, and ~~integrates chemistry and dynamics to allow simulations of the dispersionprocess, aerosol-radiation interaction, aerosol-microphysics interaction, and complex interactionsbetween chemistry, aerosols, and physics~~ other key mechanisms (Powers et al., 2017).

3.2 Model configuration

There are several gas-phase chemistry and aerosol treatments available in WRF-Chem V4.2.2. In the current study, the Model of Ozone and Related chemical Tracers Version 4 (MOZART V4) gas-phase mechanism (Emmons et al., 2010; Knote et al., 2014) is used in combination with the MOdel for Simulating Aerosol Interactions and Chemistry (MOSAIC) 4-bin aerosol scheme (Zaveri et al., 2008). Four size bins (0.039-0.156, 0.156-0.625, 0.625-2.500, and 2.5-10.0 μm dry diameters) are used in the MOSAIC aerosol module for the representation of the aerosol size distribution. The PBL scheme used in our simulation is Mellor-Yamada-Janjic (MYJ) turbulent kinetic energy (TKE) scheme (Janjić, 1990, 1994) and the land-surface model scheme is the Noah Land Surface Model scheme (Chen and Dudhia, 2001). The cumulus scheme is the Grell 3D cumulus scheme (Grell and Dévényi, 2002), which performs better than other schemes (Hasan and Islam, 2018). The cloud micro-physics scheme is the Morrison ~~2-momentum~~ 2-moment microphysics scheme (Morrison et al., 2005; Morrison and Pinto, 2005). Model radiation treatment utilizes the Rapid Radiative Transfer Model for General Circulation Models short-wave and long-wave radiation schemes (RRTMG,(Iacono et al., 2008)), including the aerosol radiation feedback.

Meteorological initial and lateral boundary conditions for WRF-Chem simulation are obtained from the National Centers for Environmental Prediction (NCEP) Global Data Assimilation System (GDAS) Final analysis (FNL) at 0.25-degree spatial resolution and 3-hour temporal resolution. The initial and lateral conditions of chemical species are obtained from the Whole Atmosphere Community Climate Model (WACCM) at 0.9*1.25-degree resolution with 88 levels. ~~All the key~~ Key species associated with wildfires are included: Carbon monoxide (CO), carbon dioxide (CO2), black carbon (BC), nitrous oxide (N2O), nitrogen oxides (NOx) and so on (Mills et al., 2016). In addition, the Fire Inventory from NCAR version 2.4(FINNv2.4) is used as fire emission input for the simulation. The model simulation was conducted over ~~Canada and CONUS region from August 9th to 25th~~two nested domains: an outer domain covering the Canadian region (25-67° N, 70-140° W) and an inner domain focused on the CONUS region (25-50° N, 64-125° W). The simulation spanned August 9th-25th, 2018, ~~at 10km spatial resolution with~~ with a spatial resolution of 10km for the inner domain and 71 vertical layers from surface to TOA (top of atmosphere). More details of the model configurations are shown in table 1.

Option type	Selected option
Horizontal grid resolution	10km (550*300)
Number of vertical layers	71 (39 layers below 2km)
Microphysics scheme	Morrison 2-moment scheme (Morrison et al., 2005; Morrison and Pinto, 2005)
Short and longwave radiation	RRTMG (Iacono et al., 2008)
Land surface	Noah-MP (Chen and Dudhia, 2001)
Boundary layer scheme	MYJ TKE scheme (Janjić, 1990, 1994)
Cumulus physics	Grell 3D (Grell and Dévényi, 2002)
Aerosol feedback	Yes
Chem-opt parameter	MOZART-MOSAIC (Emmons et al., 2010; Knote et al., 2014)
Meteorological data input	NCEP
Biogenic emissions	MEGAN
Anthropogenic emissions	EDGAR-2010

Table 1. Parameterizations used in WRF-Chem model simulations

4 Methods

This section mainly described the methods we used to estimate pollution ~~change~~ changes in the Continental United States due to long-range transported smoke. We first described the method we used for filling the AOD gaps of the MAIAC satellite AOD product, then described two different methods for estimating surface PM2.5. Finally, we discuss how we assess the surface pollution change due to Canadian wildfires.

4.1 Filling the AOD gaps

To reiterate, one of the goals of the study is to estimate daily surface PM2.5 at 0.1-degree spatial resolution. AOD alone cannot provide necessary spatial coverage due to gaps in cloud cover. Therefore, we explored two commonly used methods for filling the AOD gaps. One commonly used method for daily AOD gap-filling problems is Kriging interpolation (Kianian et al., 2021; Singh et al., 2017). At the same time, the performance of Kriging interpolation degrades with increasing distances from the training points, which implies a limitation of the method in large areas of missing data (Kianian et al., 2021). Therefore, in the second method, we combine the Kriging method with outcomes from our CTM simulations to improve the AOD-gap interpolation.

4.1.1 Kriging interpolation

The ordinary Kriging (OK) method computes the estimation of an unsampled point based on the weighted average of surrounding pixels (Zandi et al., 2011). Several authors have fully described the theoretical basis of this method (Cressie, 1988; Emery,

2005), and has been proved to successfully fill in the AOD gaps for air pollution studies (Ma et al., 2014). The estimated AOD value at an unsampled location (x0) can be expressed as:

$$Z'(x_0) = \sum_{i=1}^{n} \lambda_i Z(x_i) \tag{2}$$

where i=1,2,3...n representing for the surrounding pixels, and λ_i is the kriging weight. A major factor that expresses the spatial dependence between neighboring points is the variogram (Arslan, 2012), which is defined as:

$$\gamma_h = \frac{1}{2n} \sum_{i=1}^{n} [Z(x_i) - Z(x_i + h)]^2 \tag{3}$$

where $Z(x_i)$ is the AOD value at point i and $Z(x_i + h)$ is the AOD value of other points that have a discrete distance h from point i. Previous studies have applied OK method with exponential variogram (or semi-variogram) on interpolating missing AOD values (Lv et al., 2016; Hu et al., 2019). Therefore, in this study, we use the ~~the~~ OK method with exponential variogram to obtain a first-stage gap-filling daily AOD over our study area.

4.1.2 CTM interpolation

We applied the Kriging method for interpolation in areas with sufficient AOD information~~for interpolating, whereas we used CTM interpolation if lacking~~ , while using CTM interpolation where valid AOD retrievals ~~. This is because the possible undetected small-scale fire sources in the CTM may introduce more uncertainties if we feed the interpolation with CTM outputsin the~~ were unavailable, to minimize uncertainties associated with small-scale "missingness" ~~of AOD~~of AOD. These uncertainties arise because CTM relies on fire inventories derived from satellite fire detection products, which may fail to capture small-scale fires due to the spatial resolution of satellite observations and fractional fire coverage within a pixel (Fu et al., 2020). Such undetected fire sources can lead to inaccuracies in CTM outputs. To better represent the AOD distribution in regions with small-scale "missingness," we prioritize the Kriging method over CTM interpolation. Therefore, our gap-filling method accepts kriging interpolation in regions with a small missing portion (<20%). At the same time, we feed the interpolation with CTM outputs for regions with a more significant missing portion. The details of the process are shown in figure 1.

To estimate the AOD value ~~of a location where no~~ for a location without valid MAIAC AOD~~is present, We~~ , we first select a ~~9*9-pixel~~ 9×9-pixel box (spatial resolution of 0.1-degree) ~~. Then we calculate the differences of AOD_{wrf} between the target pixeland other points. Within the selected boxregion, we count the total number of pixels that satisfied~~ centered on the target pixel. Within this box, we identify pixels that satisfy two conditions: (a) ~~have~~ having valid MAIAC AOD ~~;~~ data, and (b) ~~have a small CTM AOD difference from the target pixel (<~~having a small AOD_{wrf} difference (0.1) ~~. Suppose the number of satisfied points is less~~ compared to the target pixel. If fewer than 50 ~~points. In that case, we increase the radius of the selected box and repeat the process, while if the satisfying points are more than~~ pixels meet these criteria, the box radius is expanded, and

the filtering process is repeated. Once at least 50 ~~, we continue to~~ pixels are identified, we calculate the ratio of ~~pixels with~~ valid MAIAC AOD ~~over the total pixels~~pixels to the total number of pixels in the box, where total pixels refer to all pixels in the selected box, regardless of filtering. If the ratio ~~is larger than~~ exceeds 80%, ~~we accept the estimated AOD using~~ the ~~OK method for the target pixel~~AOD for the target pixel is estimated using the ordinary kriging (OK) method, based on the filtered pixels. However, if the ratio is ~~less or equal to~~ 80% ~~, we computed the target AOD using a geo-weighted method considering~~ or lower, the AOD is calculated using a geographically weighted regression method that considers the neighboring ratio between MAIAC AOD and AOD_{wrf}:

$$AOD(x_0) = AOD_{wrf}(x_0) * R \tag{4}$$

where R is the weighted ratio which can be expressed as:

$$R = \sum_{i=1}^{n} (\alpha * w) \tag{5}$$

$$\alpha = \frac{AOD_{MAIAC}(x_i)}{AOD_{wrf}(x_i)} \tag{6}$$

$$w = \frac{1 - \frac{distance(x_i, x_0)}{bandwidth}}{\sum_{i=1}^{n}(1 - \frac{distance(x_i, x_0)}{bandwidth})} \tag{7}$$

Where $distance(x_i, x_0)$ means the distance between location x_i and x_0, and the bandwidth is selected based on the maximum distances between point x_i with valid MAIAC AOD and target pixel x_0 within the pre-selected box. According to above equations, we can obtain the AOD prediction for the unsampled location x_0.

4.2 Estimating surface PM2.5 using the gap-filled AOD

Having the gap-filled AOD data, we can now estimate the surface PM2.5 with more extensive spatial coverage. In this study, we tested two methods (GWR and RF) for predicting surface PM2.5 using the gap-filled AOD with other meteorological variables. Due to ~~wildfires' regional characteristics~~ characteristics of the regional wildfires, we chose both methods to consider spatial variations of pollution distribution. After comparing the fitting and validation results of the two methods, we apply the method with the better performance to estimate daily surface PM2.5.

Before we perform the prediction, different data sets need to be resampled to the exact grid resolution. Grids with ~~0.1-degree~~ 0.cru1-degree spatial resolution are constructed over the study region. For AOD data with 1km resolution, we average all valid AOD values that fall into the same grid. Moreover, for other meteorological variables with a 0.25-degree resolution, we apply the inverse distance method to scale up all the variables into the predefined grids. PM2.5 measurements in the same grid are averaged to one value to obtain a 0.1-degree resolution. In order to derive the relation between PM2.5 and AOD, we select data where AOD and surface PM2.5 are both available (AOD > 0 AND PM2.5 $> 2.0 \mu g m^{-3}$) to train the models.

4.2.1 GWR method

To derive the surface PM2.5 using filled AOD with other meteorological variables, we used a GWR model that we fully describe in our prior work (Xue et al., 2021). The GWR model has advantages over other methods because it estimates spatially varying relationships. The disadvantage of GWR model, on the other hand, is that the coefficients change daily according to different spatial characteristics of surface pollution, indicating ~~high~~ increased computational expenses. To account for varying degrees of freedom centered on different locations, Adaptive bandwidth, selected by the Akaike's information criterion (AIC), is used for the GWR model. The model can be described as:

$$PM_{2.5,i,t} = \beta_{0,i,t} + \beta_{1,i,t}AOD_{i,t} + \beta_{2,i,t}BLH_{i,t} + \beta_{3,i,t}T2M_{i,t} + \beta_{4,i,t}U10M_{i,t} + \beta_{5,i,t}RH_{i,t} + \beta_{6,i,t}SP_{i,t} + \epsilon_i, t \quad (8)$$

Where i represents different locations, t for different days, and β stands for the weight coefficients for different variables. The value of β depends on the geographical weighting of surrounding observations within the bandwidth. The weighting of each observation point decreases according to an exponential curve as the distances from the target point increase.

To test the GWR model, we must preserve only a small portion of the data, leaving most data to train the model since GWR requires an adequate number of samples, and the distribution of ground observations is uneven across the nation. Leave-one-out cross-validation (LOOCV) can be a relatively accurate way to test the model, but it requires much computational cost (Xue et al., 2021). Additionally, k-fold cross-validation with large fold numbers shows similar results (Xue et al., 2021). Therefore, we performed 100-fold cross-validation to evaluate the model performances. The entire inputs for the model are split into 100 subsets, and each time, we use one subset as testing samples while the other 99 subsets as fitting samples, after repeating this process 100 times until we test the whole data set. Finally, we evaluate the model performance by comparing the correlation coefficient (R), and root mean squared error (RMSE) of model fitting and cross-validation.

4.2.2 Random Forest method

Random forest is a non-parametric model that conducts estimations of prediction values by constructing a large number of decision trees. RF randomly divides nodes into sub-nodes for each tree, and the average estimation of different trees makes up the final results (Jiang et al., 2021a; Breiman, 1996). Of various machine learning methods, RF usually outperforms other machine learning methods due to its simplicity, diverse applications, tackling with complex cross-sensitivities among various features (Gupta et al., 2021; Jiang et al., 2021a; Zimmerman et al., 2018). The two vital parameters that affect the model performance is the tree number in the forest and feature numbers. We use 100 trees and six features in this study.

For our study period, a total of 11,942 samples are collected for training the model. The number of variables is the same as inputs for the GWR model in order to maintain consistency (AOD, BLH, T2M, U10M, RH, SP, latitude, longitude and day number). We also performed the same cross-validation as the GWR model to test the model performance so that the prediction accuracy can be compared between the two models. The model that performs better is selected to estimate the surface PM2.5 for locations where no PM2.5 observations are available based on: (a)the results of the cross-validation and (b)the differences

of results (RMSE and correlation coefficients-R) between model fitting and validation. Note that a lower RMSE and higher R indicate high prediction accuracy, and a slight difference in model fitting and validation means the model is not over-fitting.

4.3 Assessing the pollution increase caused by Canadian fires

The model that performs better from the previous step is used for estimating the gaps in surface PM2.5. To further assess the surface pollution change due to long-range transported smoke from Canadian wildfires, we conduct a control run of WRF-Chem with all fire emissions within Canada turned off while emissions in the US are kept the same. The simulated ~~AOD~~ surface PM2.5 of the control run (hereafter ~~$AOD_{control}$)~~ $PM_{2.5,control}$) and Canadian fire run (experiment run, $PM_{2.5,experiment}$) is then converted to surface PM2.5 based on the relation derived from previous steps:

$$PM_{2.5,control} = AOD_{control} PM_{2.5} * \frac{PM_{2.5}}{AOD_{filled}} \frac{PM_{2.5,WRF,control}}{PM_{2.5,WRF,experiment}} \quad (9)$$

where ~~AOD_{filled} is the full-coverage gap filled AOD, and PM2.5 is the~~ $PM_{2.5}$ is the estimated PM2.5 concentration using the better performed model (GWR or RF). The differences between $PM_{2.5,control}$ and $PM_{2.5}$ are then calculated to quantify the pollution change due to long-range transported smoke.

5 Results

5.1 Smoke plumes from Canadian wildfires transport to the US

Figure 2 shows the AOD change due to Canadian wildfires from August 17th to 20th, 2018. The changes in AOD distribution with and without Canadian fires vary according to wind directions and fire sources. The AOD change during the study period varies between 0 and 2.2 based on the two WRF-Chem simulations. The change is most prominent in North Dakota and Minnesota on August 17th, and the Canadian smoke continued to move southward to Iowa on August 18th. In the meantime, a large amount of Canadian smoke increased (high AOD increase) in Northwestern US (including Washington and Montana). On August 19th, Canadian smoke over the northwestern US was transported eastward, and more smoke was brought to the central US. On August 20th, Canadian smoke moved further to southern regions due to a storm system while the AOD values decreased, ~~which is~~ due to precipitation.

~~Compared to the previous study, a much more significant AOD change (larger high AOD coverage areas) is found in the mid-eastern US caused by long-range transport smoke from Canada (Yang et al., 2022). The larger smoke coverage areas are likely associated with larger fire emissions and beneficial transport conditions. Long-range transport smoke from Canada identified based on the aerosol height information contributes~~ Canadian wildfire smoke has been consistently identified as a contributor to AOD increases across the US during wildfire seasons. For example, during a 2015 Canadian wildfire event, AOD increases exceeding 1 were observed along the eastern US coast (Yang et al., 2022). In a 2016 event, aerosol height data revealed that Canadian smoke contributed 40%~~-60% fraction to ~~60% of~~ the total column AOD in New York (Wu et al.,

2018). ~~The observed AOD increase for high-layer aerosol~~ Observed AOD increases for high-altitude aerosols from Canadian fires ~~ranges~~ ranged from 0.18 to 0.45 at ~~532nm~~ 532 nm in New York ~~, which matches the simulated AOD change in Eastern US from 0.1 to 0.4 (figure 2). Our estimated AOD changes from Canadian fires are lower than the study that includes the US local fires. However, they match the observed aloft-layer AOD increase in New York (Yang et al., 2022; Wu et al., 2018), which indicates a reliable quantification of AOD analysis for Canadian fires within the US using chemistry models~~during this period. These findings align with the AOD increases simulated in our study and underscore the significant role of long-range transported Canadian smoke in contributing to regional pollution during wildfire episodes.

5.2 Associations of smoke transport with synoptic scale pressure patterns

Prior studies show that the transport of smoke aerosols is usually affected by synoptic pressure patterns. High surface pressure often indicates the accumulation of surface pollution, while successive low pressure can also increase surface pollution (Chen et al., 2008). In order to investigate the relationship between synoptic pressure patterns and the smoke transport process, we compared the horizontal and vertical distribution of Canadian smoke in the US.

According to the surface pressure map ~~over the CONUS region from August 17th to 20th~~on August 17, 2018 (~~shown in figure 3~~as shown in Figure 3a), there was a ~~low-pressure system in Montana on August 17th that gradually moved southeastward. The low-pressure system stayed in South Dakota on August 19th, and a storm started in Iowa on August 20th. The spatial distribution of Canadian smoke (figure 2) is primarily affected by the pressure systems: on August 17th, the high AOD associated with Canadian smoke matched the location of~~ high-pressure system located in Ontario, and its influence extended into parts of North Dakota. This high-pressure system was moving eastward, and as it moved east, the descending air associated with the ~~cold front over the northern US. Long-range transported smoke lifted by the cold front prevents the smoke from transporting further south. Due to the same reason, the high AOD in the northwestern US on August 18th also corresponds to a cold front. Another polluted region in Wisconsin is channeled by the~~ high-pressure system inhibited vertical convection, leading to the entrapment of smoke in the lower atmosphere. Consequently, the smoke experienced dry deposition at lower altitudes, which explains the low AOD concentration over northeastern Minnesota in figure 2a. Smoke at higher altitudes tended to be redirected around the high-pressure system. Simultaneously, there was a low-pressure ~~in Ohio and~~ system situated in the North Wisconsin region, causing the smoke to move in a northeasterly direction towards this low-pressure area. On August 18th, as shown in Figure 3b, the high-pressure ~~in Wisconsin. For both August 19th and~~ system shifted to Quebec, and its peripheral influence extended to the Madison region. This presence of the high-pressure system resulted in the formation of a narrow corridor of low AOD distribution within the path of smoke transport, as illustrated in Figure 2b. On the same day, a low-pressure system was forming over South Dakota and Nebraska. The ascending air and the robust winds along the trough axis established more conducive circumstances for the transportation of Canadian smoke. This phenomenon is clearly illustrated in figure 2c, where the smoke expanded further to the south, strongly influenced by the presence of the evolving low-pressure system. By August 20th, ~~the~~ this system had transitioned eastward, enveloping the states of Iowa and Missouri, as in figure 3d. Intense rainfall and thunderstorms materialized as the low-pressure system ~~in the central US promotes the transport of Canadian smokefurther south~~rotated over the area. This precipitation served as a scavenging mechanism, effectively

eliminating the smoke through a wet deposition process. Some portions of the smoke, however, carried by the powerful winds, could potentially impact air quality in the southern regions.

Figure 4 shows the surface PM2.5 distribution from EPA stations during the four days~~. The spatial patterns of surface pollution differ from the AOD change map (figure 2) in that US local fires also impact the air quality. For example, on~~ , while figure 5 presents the surface PM2.5 dry mass contributed solely by Canadian wildfires, as derived from the difference between the Canadian fire case and the control case in WRF-Chem simulations. Comparing these figures helps distinguish the contributions of local sources and long-range transported Canadian wildfire smoke to surface pollution in the United States. If regions of high PM2.5 in Figure 4 correspond to low values in Figure 5, it suggests that pollution in those areas is predominantly caused by local sources. Conversely, if high values in Figure 4 align with elevated values in Figure 5, it indicates that Canadian wildfire smoke is a significant contributor. On August 17th, ~~severe surface pollution (~~elevated surface PM2.5 ~~/~~levels (~~>30~~ 10 μgm^{-3}) ~~occurred in the northern US~~ were observed in northern US states. However, in the difference map, areas such as Montana, North Dakota, and South Dakota exhibit low values close to zero, ~~yet only North Dakotaand Minnesota show noticeable AOD increase from Canadian smoke,~~ indicating that ~~most of the~~ surface pollution in the northwestern US ~~is caused by~~ primarily originated from local fires. ~~Though minor differences exist, Canadian smoke (figure 2)matches the~~ On August 18th, a high-pressure system positioned over Minnesota resulted in higher surface PM2.5 concentrations in that region. However, the difference map shows low values over the same area, indicating that the elevated pollution levels were primarily due to local sources. High-pressure systems typically feature calm winds and stable atmospheric conditions, which trap pollutants near the surface and hinder the transport of Canadian wildfire smoke into the region. By August 19th (Figure 4c), surface PM2.5 ~~increase. As shown in figure 2, Canadian smoke moved further south with the~~ levels increased across central US states, with high AOD concentrations noted over Utah and Illinois, likely influenced by high-pressure systems. In contrast, elevated PM2.5 levels in Iowa and Missouri appear to align with the transport of Canadian wildfire smoke. A low-pressure system extending southward facilitated the movement of smoke, reaching as far south as Missouri, thereby expanding the range of smoke transport compared to the preceding days. The impact of the low-pressure system ~~on August 19th and~~ became more pronounced on August 20th ~~compared to the prior two days. In spite of the fact that ground observations are limited in the central US, the surface pollution map (figure 4) shows the same patterns~~(Figure 4d). In the center of the cyclone, particularly in Iowa and Missouri, wet deposition processes dominated. The robust winds associated with the low-pressure system carried Canadian smoke, which was subsequently removed by precipitation, leading to no significant increase in surface pollution in these areas. However, along the outer boundaries of the cyclone, from Colorado through Kansas and Oklahoma to northern Texas, surface pollution levels noticeably increased. This increase can be attributed to Canadian smoke transported by the strong winds of the low-pressure system, resulting in heightened PM2.5 concentrations in these regions.

With low-pressure systems in the central US, both model outputs and surface observations show longer southward transport paths of Canadian smoke in the US. Take August 20th as an example; air flows of the extratropical cyclone in Iowa state substantially determine the transport direction of the Canadian smoke. To investigate smoke's horizontal and vertical transport, we take the pollution distribution map of three atmospheric layers (773hPa, 850hPa, and 900hPa) to compare the moving directions of smoke (figure 6). In upper layers (773hPa), Canadian smoke intrudes into the US from two directions: northwestern US

and northeastern US. These two smoke plumes meet in the central US at higher level and then split into two directions while transporting downward (as shown in the black circles at 900hPa map). The downward transport directions can be explained using a conceptual model of the extratropical cyclone (figure 6(d)). Canadian smoke follows the downward airflow: part of the smoke moves southwesterly away from the cyclone while the rest of the pollution moves toward the cyclone and goes through wet deposition processes.

Overall, the horizontal distribution and transport of Canadian smoke are closely related to ~~cold and warm fronts~~pressure systems. Surface high pressure usually facilitates the subsidence of elevated Canadian smoke pollution (with surface PM2.5 increase). In contrast, a low-pressure system tends to have the opposite effect (lifting), corresponding to longer transport distances.

5.3 ~~Associations of smoke transport with PBL height~~

~~PBL height plays a vital role in determining the relationship between AOD and surface PM2.5 as it is considered the vertical limit of dilution volume of pollutants. PBL meteorology regulates the dispersion and mixing of pollutants between the surface and free troposphere: on the one hand, during a diurnal cycle, low PBL height at night is often related to high surface pollution concentrations; On the other hand, for daily variations, PBL height is primarily governed by synoptic pressure systems which lead to changes of surface pollution (Miao et al., 2019).~~

~~Figure ?? shows the vertical distribution of long-range transported PM2.5 dry mass (from Canada), and the black line indicates the PBL height. For the northern US, where smoke aerosols are distributed higher above ground, increasing PBL height facilitates smoke intrusion and leads to higher surface pollution. PBL height grows from early morning (18UTC) to the afternoon (21UTC), and PM2.5 concentrations also increase within the PBL. Figure ?? shows the diurnal variations of one EPA station on different days. For days with long-range transported smoke (August 20th), the increasing PBL height in the afternoon corresponds to a higher surface pollution level. However, for days without obvious wildfire smoke transport (August 29th to September 1st), PM2.5 peaks in the early morning and late night as a result of human activity and PBL height, and it is the lowest in the afternoon due to the increase of PBL height (Manning et al., 2018).~~

~~The same pattern for southern US regions during the wildfire episode (figure ??) can be seen from the vertical distribution map: increasing PBL height in the afternoon dilutes the PM2.5 concentrations, and thus surface pollution decreases. Note that the selected regions have no local wildfires, and all the smoke is transported from other fire sources. From the vertical distribution of smoke at different times, the elevated smoke layer stayed above 2km level for both day and night. Hence, the fire activity pattern has little influence on the smoke intrusion process. The reason for selecting different times in a day to analyze PBL's influence on surface pollution is to avoid the difference in smoke distribution on different days.~~

5.3 ~~Associations of smoke transport with topography~~

~~The AOD-PM2.5 relationship becomes complicated in the northwestern US due to the mountainous topography (Xu et al., 2018). Figure ?? shows the vertical cross-section of the wind direction at 45 N at 18UTC over the Bighorn mountain region (Northern Wyoming) on August 16 and 19th, 2018. The plain-to-mountain Eastern wind over this region on August 16th is nearly~~

perpendicular to the mountain range, and the upslope wind transport air upward and generates vertical updrafts above the mountain peaks, which enhances the vertical mixing process. Mountain-induced vertical mixing height can reach around four times larger than flat plain regions (Lang et al., 2015). However, the upward wind prevented air exchange between the valley and eastern plain (108.5 to 109 ° W).

However, the northwestern wind on August 18th has the opposite effect on pollution transport. The wind direction changed from northwest to southwest after passing the mountain, while the wind direction above mountain height remained the same. Therefore a lee trough is formed at the east of the mountain, which lead to a decrease in surface pressure. Thus a surface convergence zone is formed, which corresponds to the noticeable PM2.5 accumulation on the lee side of the mountain.

5.3 Daily coverage of satellite AOD and simulated AOD from WRF-Chem

During the wildfire period selected in this study (17 days), we calculate the daily MAIAC AOD coverage using the number of pixels with valid AOD values divided by the total number of pixels. The AOD coverage after combining Aqua and Terra data ranges from 46% to 68% (shown in table**??**). figure 7). Analyzing daily AOD coverage is essential, as satellite-derived AOD serves as a columnar indicator of pollution with extensive spatial coverage and high-resolution data, making it a valuable predictor for estimating surface PM2.5 concentrations alongside other variables. However, it often has missing values over fire-intense regions, which can significantly impact the accuracy of predicted surface PM values. Using Model-simulated AOD in conjunction with satellite AOD can help mitigate this issue and improve predictions.

In order to show the spatial distribution of AOD coverage, we also calculate the coverage ratio of AOD of each pixel in the 17 days (shown in figure 8). The average AOD coverage for the whole study area is around 60%. For the northeastern US, 40%-70% of the days are covered by clouds, while more than 80% of the days have valid AOD values in the western US.

The model simulated 550nm AOD has a very similar distribution as the satellite AOD. Comparing MAIAC AOD with corresponding simulated AOD pixels, the correlation coefficients range from 0.3 to 0.63, and the RMSE is within the range of 0.2-0.4 (shown in table **??**). Figure **??** shows the comparison between satellite AOD and model simulated AOD on selected daysS2). Time series of the coverage and statistics are shown in figure 7. There are no clear correlation between MAIAC AOD coverage and the correlation of two AOD products, as the missing AOD areas for each day are randomly distributed affected by satellite swath coverage and cloud contaminations. Overall, simulated AOD is lower than satellite AOD potentially due to underestimation of fire emissions, especially for small-scale fires (Wiedinmyer et al., 2011). FINNv2.4 identified fire sources based on the combinations of thermal anomalies product of MODIS and VIIRS (Visible Infrared Imaging Radiometer Suite) (Wiedinmyer et al., 2011; Li et al., 2021). Adding high spatial resolution fire detection information from VIIRS increases the total burned area by 280% compared to the previous FINN version that used MODIS detection only (Li et al., 2021). However, thick smoke and cloud cover primarily affect the detection of fires (Fu et al., 2020; Schroeder et al., 2014), causing AOD underestimation in regions with missing fire detection.

The comparison between WRF simulated AOD and satellite retrieved AOD shows high spatial correlation, indicating similar smoke pathway between the model and satellite observations. We also compare the ground AERONET AOD with the model simulated values(shows in figure **??**). The correlation coefficient during the 17 days between two AOD product is 0.54, and

the RMSE is around 0.06. Similar to the comparison with satellite AOD, model simulated AOD values show ~~underestimations~~ underestamations compared to AERONET AOD.

5.4 AOD gap filling

Our gap-filled AOD increased the mean coverage from 60% to 92%, and missing values of the gap-filled AOD were mainly distributed at the edges of our study area due to limited satellite retrieval to derive accurate $AOD_{filled} - AOD_{MAIAC}$ relationships.

To illustrate the importance and differences of feeding the interpolation with CTM outputs, we choose August 13th to display the differences when fires are detected near cloud edges. Figure 9 shows the AOD distribution of the CTM-filled AOD, kriging-filled AOD, and MAIAC AOD. The south-central US region (including Texas, Oklahoma, and Nebraska) has large missing AOD areas due to clouds. The kriging-filled AOD for this region is evenly distributed with values around 0.3. However, the CTM-filled AOD shows more variations and a clear smoke transport path along the wind direction. The primary reason for this difference is some small-scale fires detected near the cloud edges in Oklahoma. According to the fire emission document (https://www.acom.ucar.edu/Data/fire/data/finn2/README_FINNv2.5_Feb2022.pdf), both 375m resolution VIIRS fire detection and 1km resolution MODIS thermal anomalies are used for estimating fire emissions. This enhancement in fire detection provides a more accurate estimation of surface pollution in the presence of clouds. Compared with the surface PM2.5 distribution of this same day, we find the same distribution pattern as our CTM-filled AOD: high PM2.5 ($> 20 \mu g m^{-3}$) distributed in central Texas all the way to Eastern Oklahoma. Therefore, CTM-filled AOD provides closer patterns as observed surface pollution distribution at regions with large cloud covers. Our results indicate the inadequacy of kriging methods in such cases. ~~Our results clearly indicate the inadequacy of kriging methods in such cases.~~

5.5 Daily PM2.5 estimation

Figure 10 shows the GWR model fitting and cross-validation results. The ~~color represents~~ colors represent the probability function that determines the possibility that the ground-level measurements and the estimations from the GWR model are equal to each other. Lighter color means a higher concentration of samples. The R and RMSE of the GWR fitting model are 0.85 and 6.2 $\mu g m^{-3}$, respectively, and the R and RMSE of the 100-fold cross-validation are 0.8 and 7.2 $\mu g m^{-3}$. The difference between model fitting and cross-validation is relatively small, which means that the model is not overfitting and the prediction accuracy is stable. The slope of the cross-validation best-fit line (solid black line) is 0.72, indicating that the model slightly underestimates the surface PM2.5, and the biases increase with increasing AOD. The reason for this underestimation is sample biases: 1) a limited number of stations in the Western US; 2) EPA station distribution in the Western US mainly concentrates in the two populated regions, showing extremely uneven distribution. Only 3% surface PM2.5 observations exceed the unhealthy limit (PM2.5 $35.5 \mu g m^{-3}$) during the 17 days, and among these stations, 62% of the observations came from stations located in Washington and California.

Figure 11 shows the results for RF fitting and cross-validation. RF model fits well with the training samples with R of ~~0.99~~ 0.84 and RMSE of ~~1.86~~6.56 $\mu g m^{-3}$, while the cross-validation results degrade significantly (R=~~0.85~~0.76, RMSE=~~6.2~~7.69

μgm^{-3}). The slope of the validation best-fit line is ~~0.7~~0.523, which is of ~~similar values to~~ lower than the GWR method. The difference between model fitting and cross-validation indicates that the model is slightly over-fitting and has limited prediction accuracy for this case. One possible reason for this could be the limited number of training samples with average daily available measurements of around 700 during the study period (Jiao et al., 2021). A space-time RF model can be utilized to enhance RF model performance (Wei et al., 2019). Compared with ~~GWR, the RF~~ RF, the GWR model showed slightly higher prediction accuracy ~~although it is less stable~~and more stability.

Though the difference in cross-validation results for RF and GWR is slight, the daily pollution variation estimated from the two methods shows completely different trends. Figure 12 shows the mean PM2.5 variation over the three highly polluted areas (EPA Region 8,9,10) during the 17-day period calculated from GWR, RF, and EPA ground stations. For region 8, EPA stations measured two pollution peaks during the study period: one peak on August 19th and another smaller peak on August 24th. Mean PM2.5 from the RF method also captured the two peaks on the same day, while the regional mean values were slightly lower than the measurements. However, GWR has a different peak on August 11th but no noticeable increase for the ~~rest~~ remaining days. For EPA Region 9, both RF and EPA stations show a decreasing trend for the first few days and then slowly increase with time, while GWR has two clear peaks, which are not shown for the ~~rest~~ other two methods. For the most polluted region 10, the highest peaks from August 19th to 22nd are shown in EPA and RF method, but GWR shows low values for the same period. The differences between GWR and RF methods come from the distribution of sample points. ~~Most samples in EPA region ten are distributed~~ In Region 10, most EPA stations are located in Washington state, ~~which indicates that pollution increase in Washington can easily lead to an increase for the whole region 10 when calculating the~~ meaning pollution increases there strongly influence the regional mean PM2.5 ~~for the region. At the same time, the GWR method sometimes captures the variations for other regions though only limited measurements are available. Evenly distributed samples may increase the estimation accuracy a lot~~ when using RF. Conversely, GWR sometimes captures variations in other areas, even when station coverage is sparse. RF is more sensitive to the spatial distribution of ground stations, with its regional mean values closely aligning with EPA station trends. In contrast, GWR is more effective at detecting PM2.5 variations in areas with limited station coverage. An even distribution of samples could significantly improve estimation accuracy for both methods. ~~According to the variation trends of these polluted areas, we choose RF to estimate the~~ Given the observed pollution trends across these highly affected regions, GWR was selected to estimate surface PM2.5 in this study.

Our ~~RF~~ GWR daily mean estimation of surface PM2.5 during the 17-day wildfire event for each EPA region ranges from ~~3.3 to 62~~ 3.1 to 60.2 μgm^{-3} (shown in table 2). The mean surface pollution is highest at region 10 while lowest at region 1, which is comparable with previous study (Xue et al., 2021).

Date	Region1	Region2	Region3	Region4	Region5	Region6
~~Aug-9 7.6 8.9 8.2 7.1 12.2 9.3 11.6 20.9 28.5 31.0~~ Aug-10	~~7.0~~ 6.9	~~9.6~~ 7.5	~~9.7~~ 7.4	~~7.6~~ 7.5	~~14.0~~ 9.1	~~10.5~~ 10.6
Aug-11	~~6.4~~ 10.5	~~8.0~~ 11.7	~~9.4~~ 13.3	~~8.8~~ 9.7	~~16.8~~ 10.3	~~11.2~~ 9.1
Aug-12	5.1	5.8	6.9	~~9.1~~ 6.8	~~10.4~~ 12.8	~~15.8~~ 8.9
Aug-13	~~5.5~~ 6.2	~~6.3~~ 5.9	~~8.1~~ 5.8	~~12.1~~ 7.1	~~15.4~~ 12.7	~~14.4~~ 10.6
Aug-14	~~5.0~~ 6.3	5.9	6.5	~~8.7~~ 6.6	~~12.7~~ 12.8	~~14.2~~ 10.7
Aug-15	~~7.9~~ 5.9	~~14.3~~ 5.7	~~13.4~~ 5.9	~~13.1~~ 7.0	~~13.0~~ 11.4	~~13.6~~ 11.1
Aug-16	~~9.8~~ 3.1	~~15.7~~ 6.4	~~14.2~~ 8.8	~~11.0~~ 10.1	11.2	11.3
Aug-17	~~10.8~~ 5.5	~~14.1~~ 6.9	~~12.9~~ 8.9	~~8.2~~ 12.8	12.7	~~10.1~~ 10.2
Aug-18	~~6.4~~ 11.3	~~8.8~~ 11.6	~~7.8~~ 9.6	~~7.0~~ 13.2	~~14.6~~ 9.1	~~9.5~~ 11.5
Aug-19	~~6.3~~ 14.5	~~7.8~~ 12.2	~~8.3~~ 10.9	~~7.6~~ 13.2	~~12~~ 13.4	~~9.5~~ 11.3
Aug-20	~~5.9~~ 7.1	~~7.7~~ 8.3	~~7.6~~ 8.3	~~7.1~~ 7.5	~~13.2~~ 11.9	~~14.5~~ 10.2
Aug-21	~~5.8~~ 6.9	~~6.4~~ 8.7	~~6.9~~ 9.8	~~7.6~~ 9.3	~~12.0~~ 14.1	~~12.6~~ 12.2
Aug-22	~~3.3~~ 5.1	~~7.2~~ 6.4	~~8.5~~ 8.3	~~9.8~~ 9.0	~~10.8~~ 16.2	~~12.0~~ 12.4
Aug-23	~~5.8~~ 5.3	~~8.1~~ 5.5	~~9.0~~ 8.2	~~11.5~~ 10.3	~~12.1~~ 15.9	~~9.2~~ 13.9
Aug-24	~~11.8~~ 6.0	~~11.1~~ 6.0	~~9.0~~ 7.0	~~11.3~~ 10.9	~~9.5~~ 15.5	~~8.8~~ 14.8
Aug-25	~~11.4~~ 5.3	~~9.7~~ 6.6	~~10.0~~ 8.1	~~11.7~~ 12.1	~~11.3~~ 15.1	~~9.1~~ 13.5
Aug-26	7.8	13.0	13.4	14.5	13.9	13.5

Table 2. Mean PM2.5 concentrations (μgm^{-3}) over different EPA regions (1-10) estimated using ~~RF~~ GWR method

5.6 Pollution change due to long-range transported smoke from Canada

~~By applying the~~

Using coefficients derived from ~~estimated~~ simulated PM2.5 ~~and WRF-Chem AOD on AOD values simulated from the WRF-Chem control run~~in control and experiment scenarios, we estimated the PM2.5 increase ~~from~~ attributed to Canadian wildfires. Table 3 ~~shows~~ presents the daily mean PM2.5 increase ~~for~~ across different EPA regions. The regional mean PM2.5 ~~increases range from 0.001 to 7.7 μgm^{-3}, and the ratio of total PM2.5 from~~ increase reached up to 28 μg/m^3, with Canadian wildfire smoke ~~ranges from 0.02% to 13%~~contributing as much as 87.8% of total PM2.5. Region 10 ~~is most affected by Canadian fires, while region 6 is the least . The ratio of Canadian smoke-induced~~ experienced the highest impact, while Region 1 was the least affected. Notably, on some days, the contribution of Canadian wildfire smoke to total PM2.5 in ~~Northeastern US (regions~~ the Northeastern U.S. (Regions 1, 2, and 3) ~~can be even larger than in northwestern US (regions~~ exceeded that in the Northwestern U.S. (Regions 8 and 10)~~on some days. During the wildfire episode, the variations of the ratios followed the wildfire activity pattern. However~~. The contribution of wildfire smoke to total PM2.5 closely followed wildfire activity patterns; however, due to ~~the~~ transport distances, it ~~usually took about 24 hours for the smoke to move from region~~ often took

time for smoke to travel from Region 10 to ~~region~~ Region 1. ~~This is why the~~ As a result, during some wildfire episodes, PM2.5 ~~change pattern in Eastern CONUS is often a day behind Western CONUS~~changes in the Eastern CONUS lagged behind those in the Western CONUS by a day.

Due to the CAA, the overall trend of surface PM2.5 in the recent years is gradually decreasing (figure ~~??~~S1). The decreasing trend is evident in August mean PM2.5 calculated from EPA surface observations from EPA region 1 to region 7, but variations of PM2.5 in region 8 to 10 is more affected by the occurrence of wildfires. For years with low wildfire occurrences (FRP $<$ 10000 ~~MW~~MJ), August surface PM2.5 show a steady pattern in region 8 and region 10 without any apparent rising or dropping. PM2.5 in region 9 shows descending pattern in August in years without large fires, while it can reach up to three times more of the baseline in wildfire years. For regions 1 to 7, August mean PM2.5 concentrations decrease about 5% -10% each year in different regions. Compared with table ~~??,~~ S1, the 17-day investigation highlights how long-range transported smoke from Canada ~~compensates for~~ temporarily offsets the descending trend in ~~a negative fashion. For region~~ surface PM2.5 during the study period. For regions 8 to 10, wildfires (~~combining~~ including contributions from both local and remote fires) ~~increases~~ increase the August mean surface PM2.5 by ~~7%- 16%.~~ 0-98%. While this study focuses on a short-term event, it demonstrates the significant seasonal impact of Canadian smoke on air quality, emphasizing the need for multi-year investigations to assess long-term trends in Canadian smoke contributions.

In conclusion, due to the concurrent local and remote wildfires, the long-range transport smoke contributed to about half of the surface pollution increase in EPA regions 8, 9, and 10. For other EPA regions, Canadian smoke compensated the CAA, causing ~~pollution decreasing trend, and causing~~ surface pollution to rise.

Date	Region1	Region2	Region3	Region4	Region5	Region6	Re
~~Aug-9 6.8 6.7 9.7 8.3 8.9 10.0 9.3 7.0 5.6 9.4~~ Aug-10	~~6.1~~ 34.3	~~10.7~~ 39.1	~~11.2~~ 39.1	~~9.1~~ 43.9	~~4.3~~ 59.9	~~3.8~~ 64.3	~~8.5~~
Aug-11	~~1.7~~ 8.5	~~3.5~~ 15.8	~~3.6~~ 16.7	~~6.5~~ 19.4	~~4.7~~ 0	~~3.9~~ 26.1	~~5.4~~
Aug-12	~~8.6~~ 0	~~6.8~~ 0.1	~~4.0~~ 0.5	~~3.8~~ 6.7	~~4.0~~ 21.0	~~4.4~~ 28.8	~~4.5~~
Aug-13	~~0.2~~ 1.7	~~0.02~~ 6.0	~~0.3~~ 8.5	~~1.9~~ 7.7	~~4.4~~ 17.6	~~2.7~~ 4.1	~~3~~
Aug-14	~~6.0~~ 1.0	~~5.9~~ 0.7	~~3.9~~ 0	~~3.0~~ 0	~~4.5~~ 5.0	1.7	
Aug-15	~~2.6~~ 4.9	~~3.4~~ -0.6	~~5.5~~ -6.9	~~3.5~~ -2.5	~~3.1~~ -2.1	~~1.0~~ 0.1	~~2.~~
Aug-16	~~3.3~~ 48.8	~~4.0~~ 40.6	~~3.4~~ 41.9	~~2.1~~ 51.6	~~6.8~~ 67.2	~~2.8~~ 62.9	~~4.~~
Aug-17	~~7.7~~ 65.0	~~8.0~~ 68.4	~~5.4~~ 74.3	~~3.9~~ 77.7	~~5.0~~ 81.5	~~3.4~~ 71.6	~~3.~~
Aug-18	~~3.1~~ 62.8	~~3.9~~ 60.9	~~3.3~~ 61.0	~~2.4~~ 66.6	~~7.0~~ 69.7	~~1.7~~ 67.3	~~5.~~
Aug-19	~~9.8~~ 57.1	~~8.4~~ 59.6	~~7.7~~ 62.9	~~4.5~~ 64.0	~~10.8~~ 66.7	~~4.4~~ 66.3	~~6.~~
Aug-20	~~8.6~~ 13.0	~~7.0~~ 23.0	~~3.5~~ 24.1	~~4.0~~ 24.1	~~4.7~~ 18.9	~~3.3~~ 33.3	~~1.~~
Aug-21	~~0.3~~ 23.6	~~5.5~~ 19.9	~~4.8~~ 26.3	~~2.1~~ 26.7	~~11~~ 26.3	~~4.3~~ 14.8	~~8.~~
Aug-22	~~12.2~~ 52.6	~~11~~ 60.2	~~9.5~~ 64.3	~~8~~ 69.1	~~10~~ 59.1	~~6~~ 73.5	~~8.~~
Aug-23	~~11.6~~ 62.0	~~13.1~~ 63.6	~~6.3~~ 75.6	~~9.1~~ 80.1	~~5.9~~ 87.4	~~2.5~~ 85.6	~~6.~~
Aug-24	~~6.1~~ 66.4	~~5.1~~ 69.5	~~7.5~~ 71.9	~~6.1~~ 79.1	~~6.5~~ 69.6	~~10.6~~ 76.1	~~9.~~
Aug-25	~~12.6~~ 55.4	~~9.3~~ 56.2	~~5.5~~ 54.6	~~5.2~~ 53.9	~~6.5~~ 56.4	~~7.3~~ 62.5	~~5.~~
Aug-26	52.3	51.6	59.5	67.3	49.2	67.6	

Table 3. Percentage increase of PM2.5 concentrations (%) due to Canadian wildfires over different EPA regions (1-10) estimated using ~~RF~~ GWR method

5.7 Uncertainties and limitations

The main uncertainties and limitations of this study come from the CTM model and various inputs of the model, and the surface pollution estimation model also leads to some uncertainties:

1) Since satellite fire detection is affected by various factors, including cloud cover, fire sizes and the background environment, emission inputs for the WRF-Chem simulations derived from the fire detection products ~~can bring~~ introduces uncertainties into our simulations and create regional biases in the simulated AOD values.

2) ~~Despite~~ In addition to the fire detection biases, ~~other assumptions and estimations used in calculating~~ assumption made in computing fire emissions also ~~add uncertainties to the simulation~~cause the following uncertainties that affect the simulations: a)fire sizes and duration; b)amount and distribution of biomass fuels; c)fraction of different emissions from the biomass fuel Soares et al. (2015). These factors may influence the mass concentration and distribution of smoke aerosols.

3)Uncertainties in the injection height of smoke plumes in WRF-Chem can impact simulation accuracy. Biases in injection height influence horizontal transport speed, direction, and pollution residence time, potentially introducing errors in the AOD

gap-filling process. Moreover, uncertainties in the vertical distribution of aerosols can affect the AOD-PM2.5 relationship, with variations in plume height between monitors leading to inaccuracies in estimated surface PM2.5 concentrations.

4) The unevenly distributed EPA stations primarily affect the performance of the two pm2.5 estimation models, causing completely different daily variation trends of regional mean PM2.5. Therefore, the model performance may be improved if we use more EPA stations (We use FRM monitors only in this study).

45) Since we need the relationships between satellite AOD and CTM AOD to calculate the filled AOD values, the filling values cannot be derived if the area of missing satellite AOD is larger than the radius thresholds we set for deriving the relationships. For days with large areas of missing satellite AOD at the boundary region of our study region, we sometimes have missing AOD values at the boundary. This can be improved by increasing the radius thresholds or the study region to leave space for the boundaries.

6 Conclusion

This study first analyzed the influence of different physical processes on the transport of long-range transported smoke aerosols by comparing two WRF-Chem simulations with and without Canadian wildfires. Then we utilized the simulated AOD from CTM and Kriging interpolation with a geographically weighted method to fill in the daily AOD retrieval gaps caused by cloud covers. ~~After that,~~ Then we estimated the surface PM2.5 concentration using GWR and RF methods and tested the two predictions using cross-validation and trend analysis to choose a better-performing method. Finally, by turning off the Canadian wildfire emissions in the CTM simulations, we calculated the surface PM2.5 concentrations from the CTM AOD outputs using the coefficients derived from previous estimations. The differences in PM2.5 of the two estimations indicated the change brought by long-range transported smoke from Canadian wildfires. The main findings of our study are:

1) Synoptic scale pressure systems are the dominant drivers of horizontal pollution transport pathways. In the meantime, the pressure systems can also affect the vertical distribution by ascending or descending smoke. Under most circumstances, the subsidence flow of high-pressure systems facilitates the drifting process of elevated smoke layers and thus increases surface pollution. In contrast, the cyclonic storm system leads to a longer transport path (further south of CONUS in this study) and directs the elevated smoke to the ground in different directions. Therefore, the co-occurrence of low-pressure systems and smoke aerosols corresponds to larger pollution areas.

~~With favorable transport conditions, certain regions may suffer from severe air quality degradation from long-range transported smoke. During the fire season, the Canadian smoke in combination with the western US wildfire smoke counteracts the CAA and post severe threats to public health across the nation. The wildfire smoke can not only degrade ground-level air qualitybut also increase atmospheric and precipitation acidity and further affect the local ecosystem if the frequency and intensity of wildfires continue to increase in the future (Boy et al., 2008; Levine, 2003). Therefore, it is necessary to improve the predictions of wildfire occurrence and transport pathways in order to warn the public in time.~~ Certain weather patterns can facilitate the dispersion of smoke, hence, during severe wildfire events, it becomes crucial to closely monitor the meteorological conditions and alert the public about the potential risks of deteriorating air quality, even if the fires are hundreds of miles

away. Furthermore, when conducting prescribed fire burning, it is essential to take into account the possibility of smoke being transported to densely populated areas. In other words, proper consideration of the weather conditions and potential wind patterns is crucial to minimizing the impact on nearby populations.

~~2) PBL height negatively correlates with surface pollution in most cases with pollution sources below the boundary layer. In contrast, it positively correlated with surface pollution when the bottom of the long-range transported smoke layer drifted close to the boundary layer.~~

~~3)Through analyzing the vertical motion over the Bighorn mountain region, we found that the vertical distribution of pollution is primarily affected by terrain topography and wind directions. Westerly winds may cause pollution accumulation in the eastern plain, while easterly winds can enhance vertical mixing, decreasing pollution over the eastern plain.~~

~~4~~2) Daily AOD coverages combining Aqua and Terra MODIS range from 46% to 68% during our study period, and our filled AOD values using CTM AOD outputs are able to fill in the missing gaps~~, and serve as bases for daily PM2.5 estimations~~.

~~5~~3) Daily PM2.5 estimations using the filled AOD product with other meteorological data using the ~~RF~~ GWR method (R=~~0.85~~0.8) perform better than the ~~GWR~~ RF model (R=~~0.8~~0.76), and the RF model captures the daily variations of different EPA regions calculated from EPA stations.

~~6~~4) Regional mean ~~ratio of total~~ increase of surface PM2.5 concentrations that came from Canadian wildfire smoke ranges up to ~~13%~~ 28 μgm^{-3} (a 69% increase), and EPA Region 10 is most affected by Canadian fires while region 6 is the least. The PM2.5 change pattern in Eastern CONUS often lags the Western CONUS by a day.

Our study found that ~~specific weather systems can enlarge the areas~~ presence of synoptic scale pressure systems leads to a higher proportion of CONUS region being affected by long-range transported smoke from Canada. ~~Besides, the surface pressure systems dominate the smoke transport paths. This information can be helpful for vulnerable populations in avoiding exposure to smoke pollution. Without the help of meteorological conditions, Canadian smoke can only influence limited regions in the US; however, with specific low-pressure systems, the pollution may pose serious health threats to much more~~ Typical airflow patterns that are associated with extratropical cyclones are particularly effective at transporting elevated layers of smoke to the surface and fanning the associated particulate pollution over large areas. Such transport pathways associated with extratropical cyclones need to be considered when forecasting for the effects of smoke pollution from Canadian wildfires on vulnerable populations in the ~~US. In addition, the weather system usually comes with large cloud covers, which restrict satellite retrievals; therefore, CTM simulations are significant for predicting surface pollution in higher spatial and temporal resolution. Furthermore, the daily PM2.5 estimations show air pollution rebounds in wildfire season after years of emission control. Moreover, the smoke-induced pollution increase can be much more than the industrial pollution , indicating the significance of predicting surface pollution and warning the public~~CONUS region. Our study highlights the significant contribution of wildfires to particulate pollution during the study period, aligning with prior research that suggests wildfires are becoming an increasingly important source of particulate pollution as industrial pollution declines due to stringent regulations (Xue et al., 2021). However, further multi-year investigations are needed to robustly confirm this trend on a broader temporal scale.

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Figures

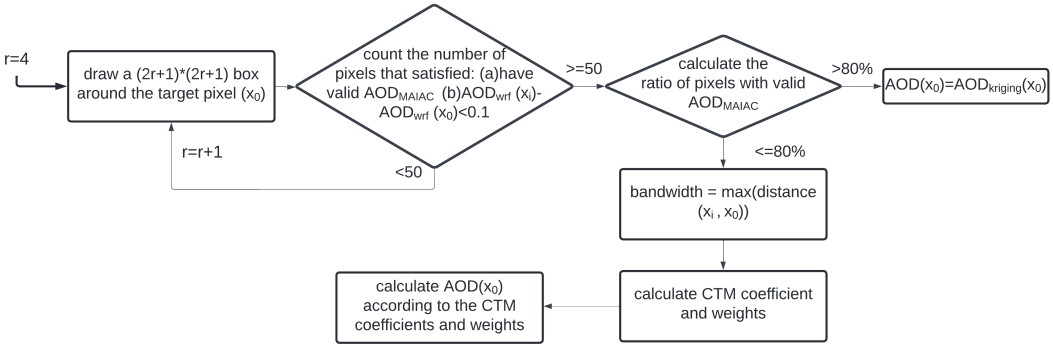

Fig. 1. ~~*flowchart of the AOD gap-filling process*~~ *flowchart of the AOD gap-filling process. "r" referse to the radius of the selected box used in the process.*

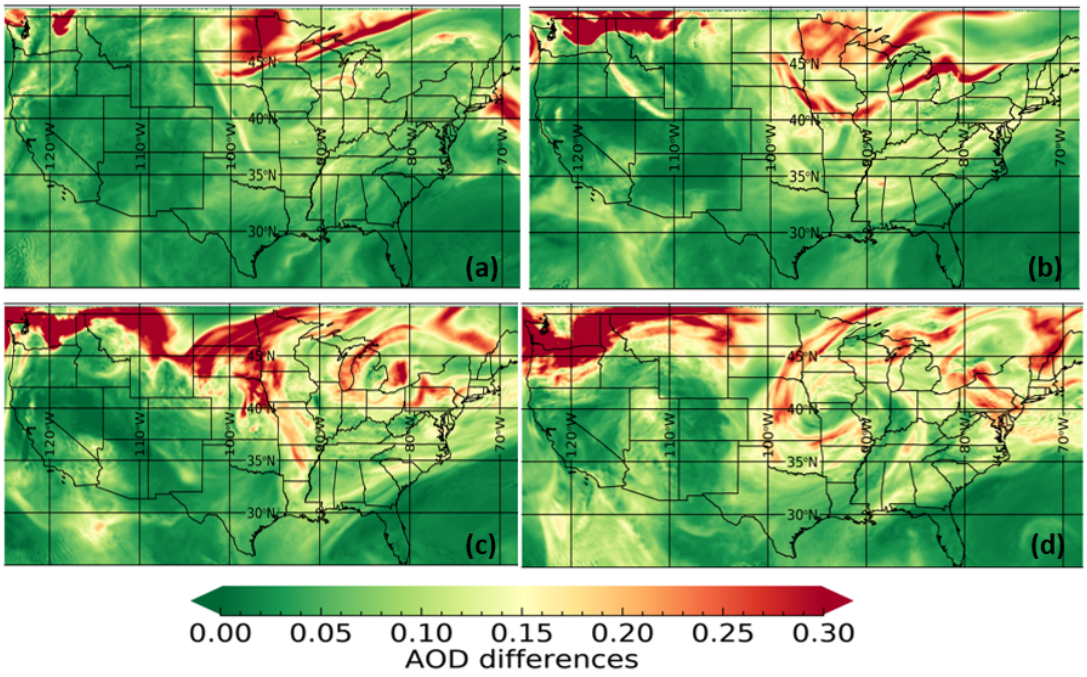

Fig. 2. ~~*AOD change due to Canadian wildfires from August 17th to 20th, 2018. The AOD change is calculated using the difference of two WRF-Chem simulations.*~~ *AOD change due to Canadian wildfires from August 17th to 20th, 2018: (a)August 17th,2018; (b)August 18th,2018; (c)August 19th,2018; (d) August 20th,2018. The AOD change is calculated using the difference of two WRF-Chem simulations.*

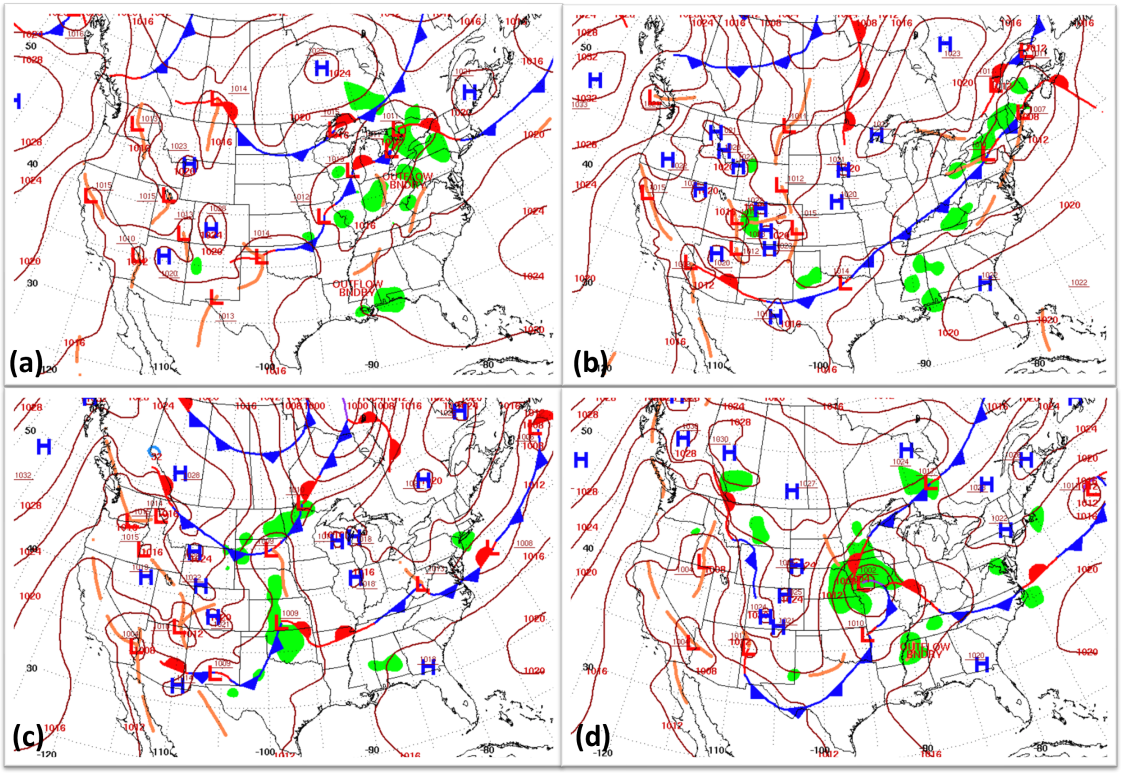

Fig. 3. ~~*Surface weather maps from August 17th to 20th, 2018 ().*~~ *Surface weather maps from August 17th to 20th, 2018 (https://www.wpc.ncep.noaa.gov/archives/web_pages/sfc/sfc_archive.php): (a)August 17th,2018; (b)August 18th,2018; (c)August 19th,2018; (d) August 20th,2018.*

~~*Simulated vertical cross section of PM2.5 dry mass with PBL height in 45 ° N (first row) and in 35 ° N (second row) at 18UTC (first column) and at 21UTC (second column).*~~

~~*Diurnal variations of surface PM2.5 concentrations from one EPA station (39.7 ° N, 104.9 ° W) on different days.*~~

~~*Simulated vertical cross section of PM2.5 dry mass with wind direction and terrain height in 45 ° N on August 16th (left) and 19th (right) at 18UTC.*~~

~~*Comparison between WRF-Chem AOD and AERONET AOD from August 9th to August 25th. The dotted line shows the 1:1 line, and the black line is the best fitting line.*~~

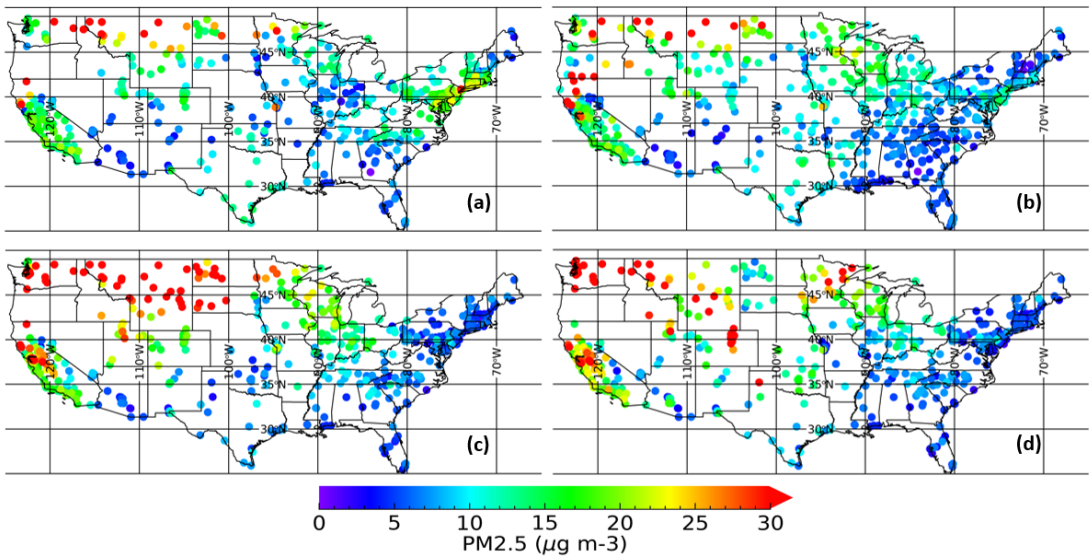

Fig. 4. ~~*Surface PM2.5 measurements from EPA stations over CONUS from August 17th to 20th, 2018.*~~ *Surface PM2.5 measurements from EPA stations over CONUS from August 17th to 20th, 2018: (a)August 17th,2018; (b)August 18th,2018; (c)August 19th,2018; (d) August 20th,2018.*

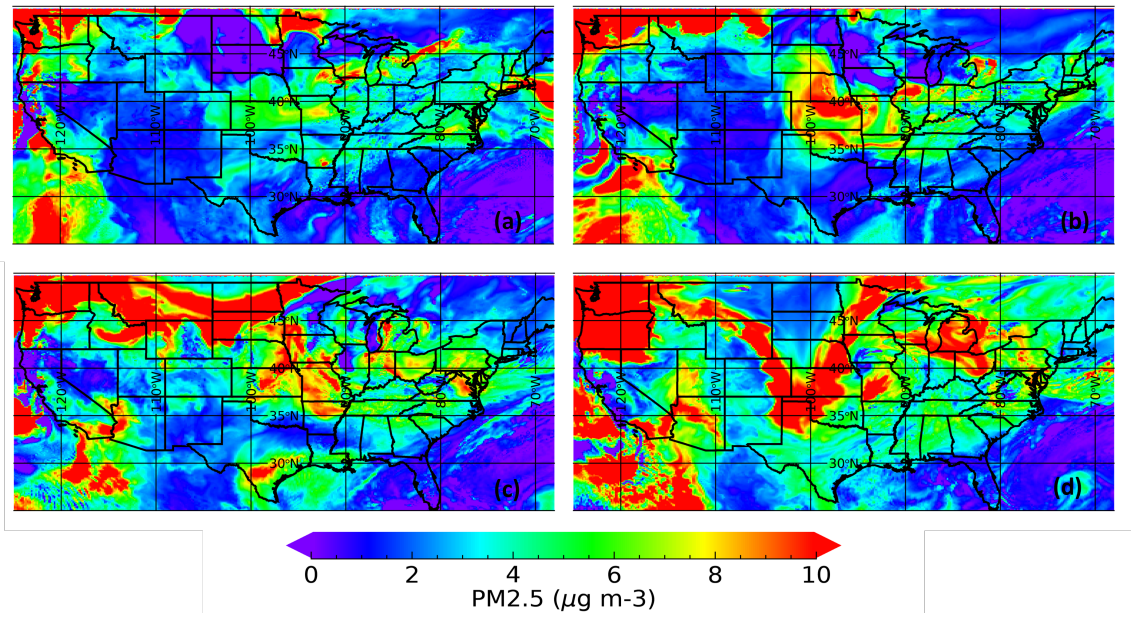

Fig. 5. ~~*PM2.5 dry mass distribution on August 20th,2018 at different levels (a)773hPa (b)850hPa (c)900hPa (d)conceptual model of the extratropical cyclone around Iowa state.*~~ *Surface PM2.5 dry mass difference from WRF-Chem simulation over CONUS from August 17th to 20th, 2018: (a)August 17th,2018; (b)August 18th,2018; (c)August 19th,2018; (d) August 20th,2018. The PM2.5 difference is calculated using the two WR-Chem simulations.*

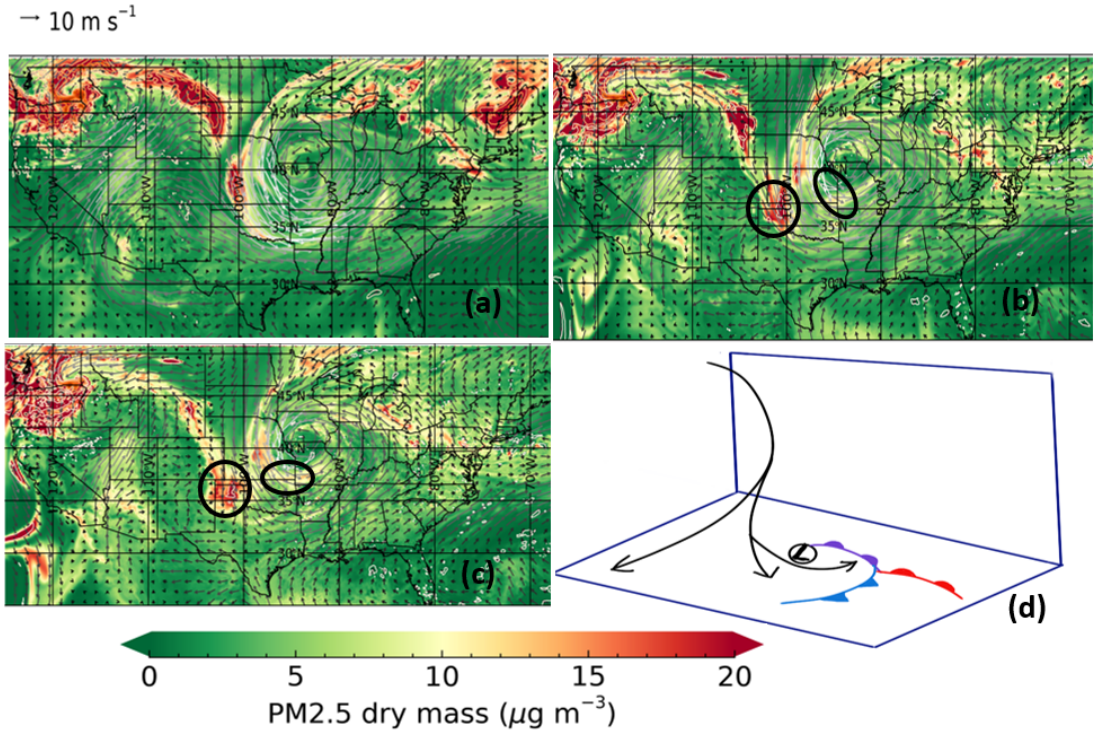

Fig. 6. *PM2.5 dry mass distribution on August 20th,2018 at different levels (a)773hPa (b)850hPa (c)900hPa (d)conceptual model of airflow patterns in a typical extratropical cyclone based on (Cotton et al., 2011).*

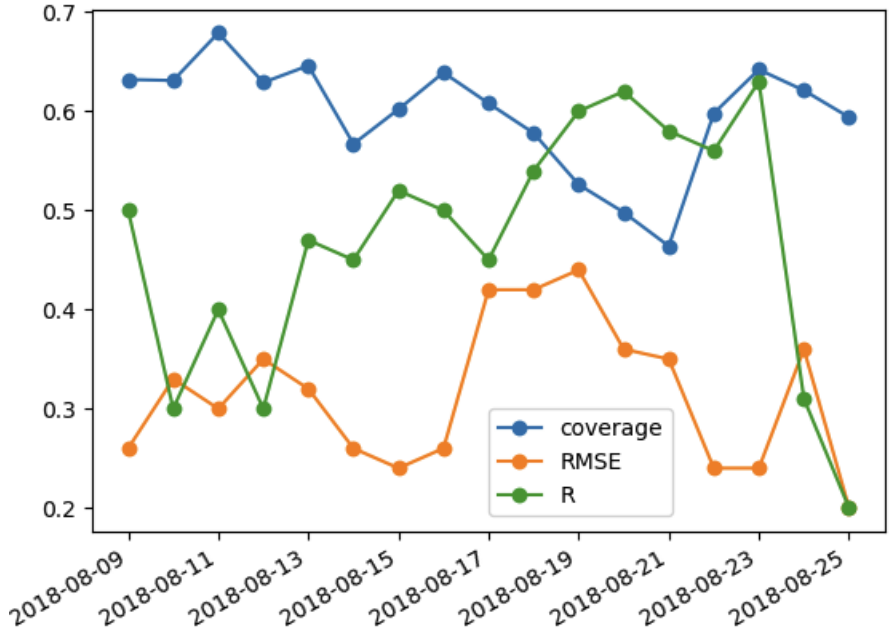

Fig. 7. ~~*AOD coverage during the 17-day period*~~ *Time seires of MAIAC AOD coverage (blue line), correlation coefficient (green line) and RMSE (orange line) of two AOD products (WRF-CHEM AOD and MAIAC AOD).*

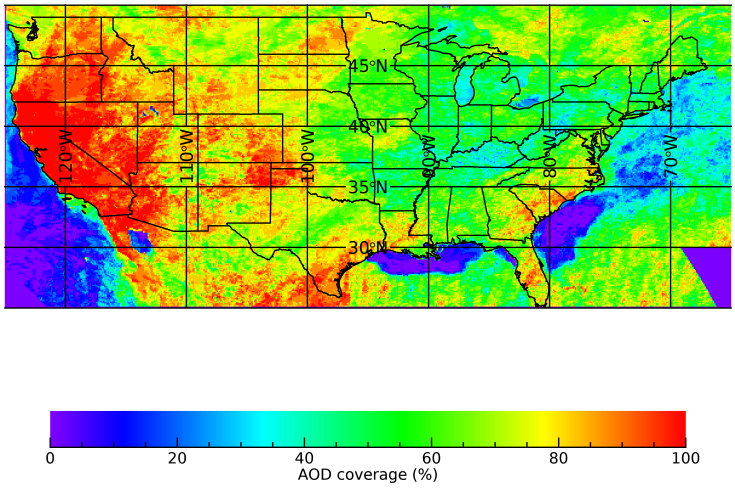

Fig. 8. ~~*Comparison between WRF-Chem AOD and MAIAC AOD on August 11th (left) and August 20th (right).*~~ *AOD coverage during the 17-day period*

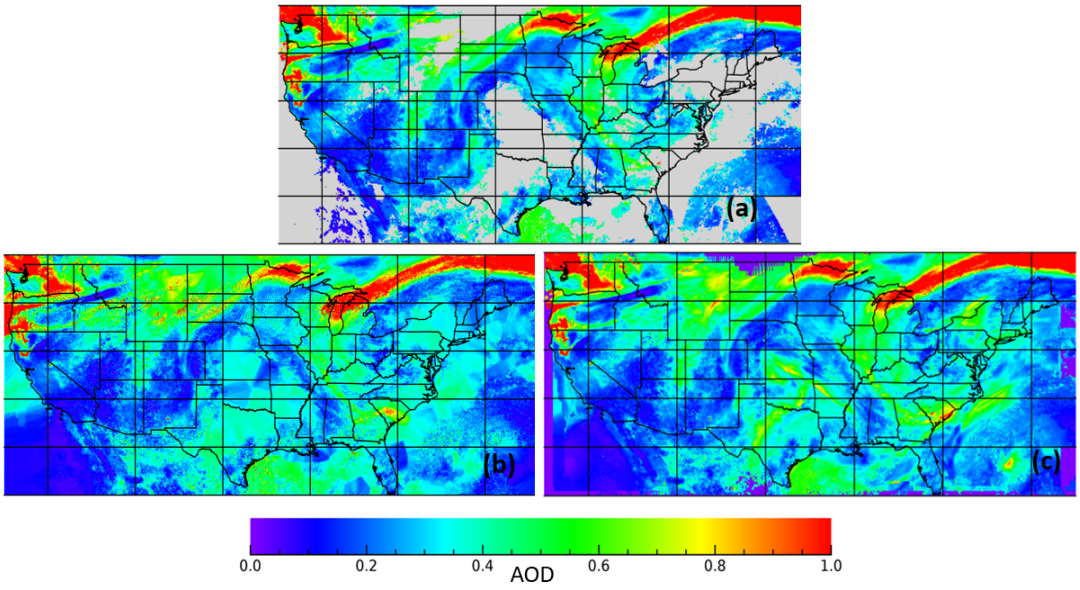

Fig. 9. ~~*AOD distribution from MAIAC (top), Kriging method (lower left) and CTM-filled AOD (lower right) on August 13th 2018.*~~ *AOD distribution from (a) MAIAC, (b) Kriging method and (c)CTM-filled AOD on August 13th 2018.*

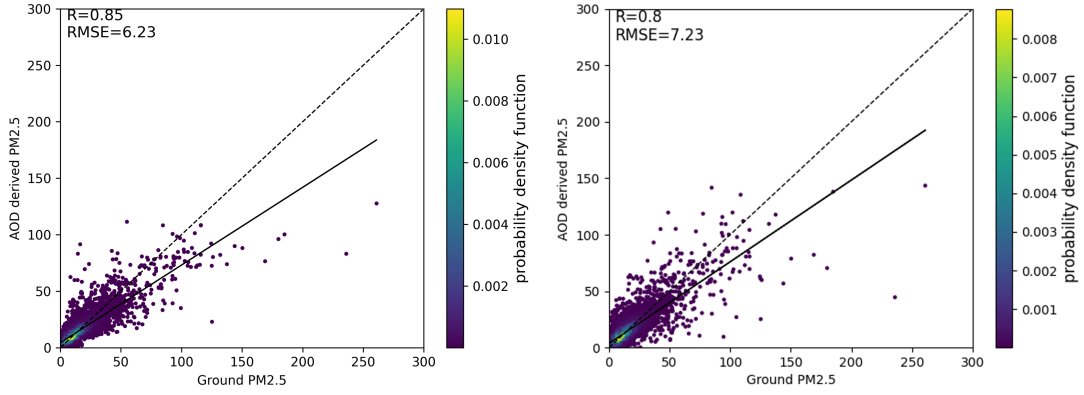

Fig. 10. ~~*Fitting and k-fold cross validation results for GWR method*~~ *Fitting and 100-fold cross validation results for GWR method*

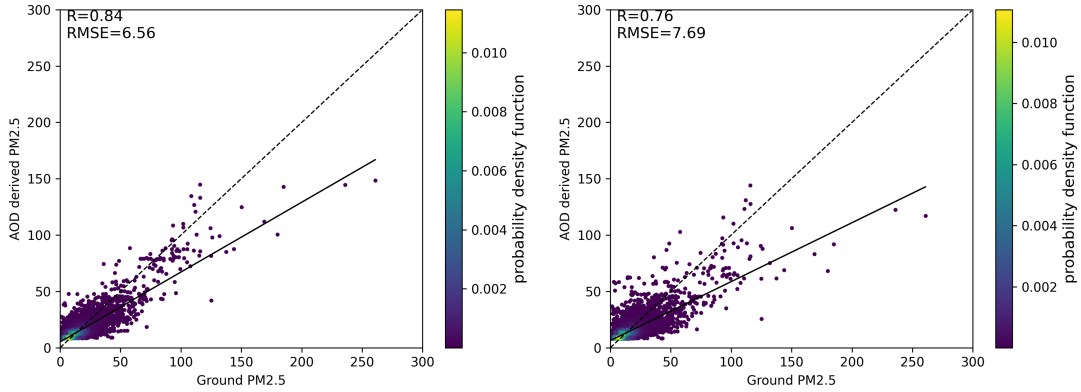

Fig. 11. ~~*Fitting and k-fold cross validation results for RF method*~~ *Fitting and 100-fold cross validation results for RF method*

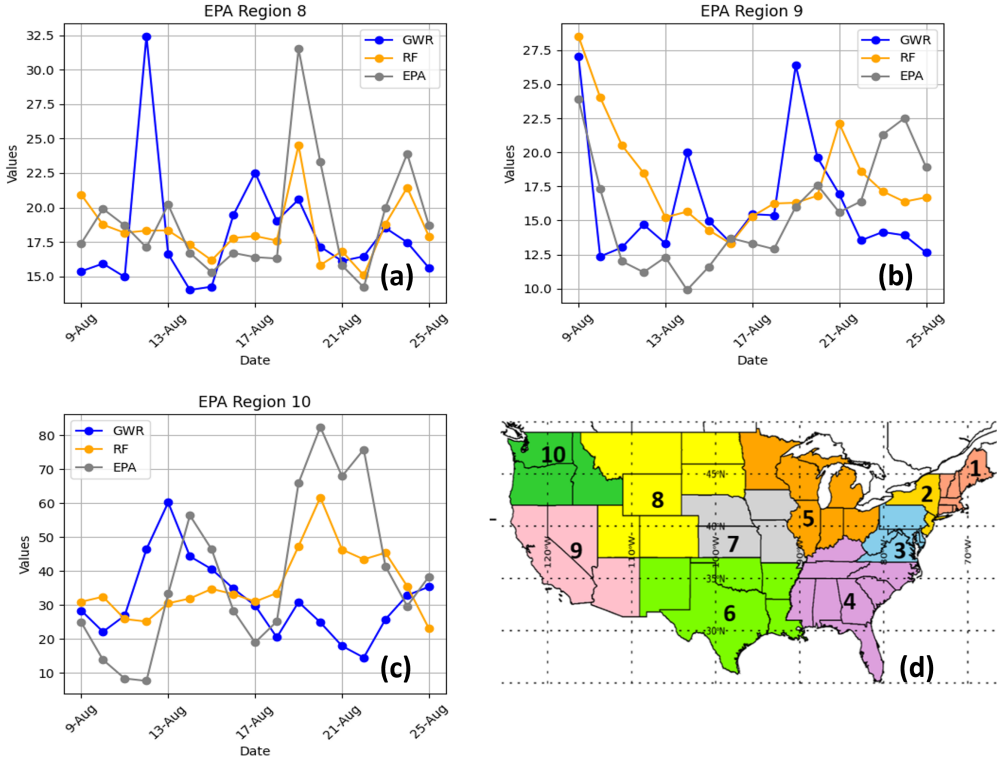

Fig. 12. ~~*Mean PM2.5 concentration variations over the top 3 polluted areas (EPA region 8,9,10).*~~ *Mean PM2.5 concentration variations over the top 3 polluted areas (a) EPA region 8 (b) EPA region 9 (c) EPA region 10 (d)EPA region map*

Appendix A: ~~Appendix A~~

~~Figure?? shows the changing trends of monthly surface PM2.5 and total FRP from January 2001 to December 2020 (a total of 240 months). There are much variations of monthly change of both surface PM2.5 and total FRP due to the inter-annual characteristics of surface PM and wildfires, and some variations between the two variables matches well with each other but some are not. Of all the regions and months, the largest FRP occurs in 236 (September 2020) at EPA region 8. The total FRP of EPA region 8 in September 2020 reaches 4675228.5 MW and the corresponding monthly mean PM2.5 is around 30 μgm^{-3}. Both surface pollution level and FRP peaks match well in this month which indicates that large fires largely increase the surface PM2.5. Large wildfires (high FRP) are commonly occur at EPA region 8, 9 and 10. total FRP larger than $5\times2\times10^5$ MW usually lead to obvious increasing/peaks of corresponding surface PM2.5. However, low FRP (small scale fires) hardly cause any obvious increase on the surface pollution levels. Of all the regions, PM2.5 at region 10 are most sensitive to FRP changes which could because of the lowest population (15.7 people per square mile) in all EPA regions. Sometimes the variation of PM2.5 is also influenced by wildfires from other regions. In August 2018, there are large wildfires in region 9 and 10 (peaks at x=211, frp=$7.3\times2\times10^5$ MW for region 9 and frp=$5.4\times2\times10^5$ MW for region 10) and no large fires in region 8, while surface PM2.5 in region 8 shows a peak corresponding to the wildfires in the neighboring region.~~

~~Over the 20 years, surface PM2.5 in EPA region 1 to region 7 are showing noticeable decreasing trend. Surface pollution in EPA region 9 also shows decreasing trend in months without large fires (low FRP), however, EPA region 8 and 10 have a much constant surface PM2.5 values in the 20 years and no obvious change in low wildfire months.~~

~~*Monthly change of mean surface PM2.5 concentrations (black) and monthly total FRP (red) in different EPA regions (EPA region 1 to 10) from January 2001 to December 2020 (a total of 240 months).*~~

~~Estimated yearly decrease in surface PM2.5 using linear regression models~~

Author contributions. ZX and SC conceived and designed the study. ZX performed the data analysis and prepared manuscript. ZX, SC and NU contributed to editing the manuscript and provided valuable suggestions on data analyss and interpretation.

Competing interests. The authors declare no competing interests.

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